Manipulating phonons at the nanoscale

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Abstract

Efficiently controlling phonon propagation is important in many applications, ranging from the design of thermoelectric materials to thermal budget engineering. Phonon manipulation, nonetheless, has proved to be an elusive task. Here we review some basic strategies to alter vibrational modes throughout the entire phonon spectrum by means of point and extended defects, boundaries, and the design of periodic superstructures. These individual scattering mechanisms allow altering the phononic properties in specific spectral regions, while combing two or more can lead to a modification of the full-spectrum.

Keywords: `phonons, phononics, nanoscale thermal transport, point defects, superlattices, thermoelectrics`

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1. Introduction

Phonons, the quanta of lattice vibrations of a solid, are the particles that carry heat in insulators and semiconductors[1,2]. Many applications in fields that range from energy harvesting to information technology would benefit from a tight control on their nature and their propagation in presence of a temperature gradient. Controlling and handling phonons, however, is an inherently challenging task: a proof of this fact is that heat is commonly regarded as a source of loss. *Phononics* is the discipline that aims at manipulating phonons, either to give a material tailor made vibrational properties or tune phonon propagation to implement logic functions[3,4].

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Recent years have witnessed an enormous progress in the growth and design of nanostructures and now materials with unprecedented level of purity and structural quality are available. Present experimental capabilities are such that nanostructured features of the same characteristic length of phonons can be obtained. This enhanced degree of control in material design opens the way to a wealth of new strategies to control and manipulate phonon transport. The thermal conductivity of a material can be purposely suppressed, to engineer an efficient thermoelectric [5]; thermal budget, which otherwise can be the bottleneck of the performance of many nanoelectronic devices, can be lowered [6]; phonons can be used to encode logic function in devices analogous to their electronic counterparts [3], such as diodes [7,8,9] and transistors [10,11], where heat propagation can be controlled thermally [10,12], electrically [13,14,15], and mechanically [16,17,18,19].

In this brief review we will discuss the most common strategies to alter the thermal conductivity and the phononic band structure with the goal of designing materials with desired vibrational properties. To this end, we will review some of the most significant and recent contributions about the role of impurity and boundary scattering and about superlattice design.

2. Impurity and boundary scattering

Tailoring the properties of materials by means of the intentional addition of defects or impurities is certainly not new and the design of electronic devices almost entirely relies on this principle, *i.e.* the juxtaposition of regions of a semiconductor with different doping characteristics [20]. This strategy can be effectively pursued to tune the amount of phonon scattering as well [21,22,23,24,25,26]. An important difference with electrons when it comes to point defects, however, is that for a phonon to be scattered the impurity does not need to have a different chemical identity from the atoms of the host lattice, since a different mass is enough. For this reason isotopes are the paradigmatic impurities in phonon transport. As matter of fact, for most of the materials found in nature or synthesized in the lab, a distribution of the isotope population is a natural feature. This fact makes isotopes a very special class of impurities, as they are unintentional defects that are not added on purpose and are always present, unless specific purification processes are followed. Indeed, when looking up the thermal conductivity of materials in datasheets the value normally reported is that of the *natural* material, *i.e.* with the natural distribution of isotope population.

In the isotope limit, where only the mass of the impurity changes, the distortion of the crystal lattice is extremely localized. Such a distortion can extend up to the first- or second-neighbor shell if also the electronic configuration changes, particularly for those impurities that favor different bonding arrangements, or in the case of vacancies. The range of the distortion of the lattice gives an estimate of the frequency of the phonons that will be scattered by the defect: long wavelength phonons will not notice a very localized perturbation of the lattice. It has been shown that isotopes of Si or BN nanotubes act like a low-pass filter [27,28]: low frequency acoustic phonons go through almost unaltered, while high frequency phonons are scattered.

Extended, disordered defects such as those encountered in form of voids in nanoporous materials are much more effective in tuning the thermal conductivity, because they are also able to effectively suppress long mean free path phonons that carry a significant fraction of the heat [29,30,31,32,33,34,35,36,37,38,39,40]. Remarkably, small porosities of less than 5% are sufficient to decrease greatly the thermal conductivity, while going above 15% only results in a further, but marginal reduction. This behavior is very similar to the one observed in SiGe alloys or in the previously discussed case of isotope disorder where the dependence of κ on the composition exhibit a characteristic U-shape [41]; see Figure 1.

Another possible approach to manipulate phonon transport is by tuning the amount of boundary scarring [42,43] that they suffer as they travel through a nanostructure [44,45]. When a phonon hits the surface of a material it is scattered. Samples are finite and thus there is always a boundary to scatter phonons, though clearly it is in systems with a reduced dimensionality and characteristic sizes in the order of phonon mean free path that the effect of boundary scattering is more pronounced. The role of boundary scattering becomes dominant especially at low temperature, where phonon-phonon scattering is much less efficient and can be neglected. In these conditions it is much more likely that a phonon reaches the surface and is therein scattered rather than colliding with other phonons and thus its lifetime will be determined by boundary scattering. With the advent of nanowires [46,47,48] and nanostructured semiconductors in general [49,50,51,52] the importance of this scattering mechanism has increased considerably and strategies to partially tune boundary scattering —by for instance engineering rough surfaces [53,54,55]— have been put forward. In their seminal work [48], recently completed by a follow-up paper where the high temperature regime was also explored [56], Li and coworkers measured the diameter dependence of the thermal conductivity in thin nanowires, the ultimate *smoking gun* of boundary scattering.

Boundaries also play a crucial role in highlighting heat transport regimes that go beyond the well-known Fourier's law, the phenomenological linear relation between heat flux and thermal gradient. Phonon hydrodynamics [57,58,59,60], where phonons travel collectively and can be described with a close analogy with fluid dynamics, can be observed in thin nanowires. There the reduction of thermal conductivity close to the boundaries leads to a curved profile of the heat flux, which is larger near the axis of the wire larger than on the wire boundary.

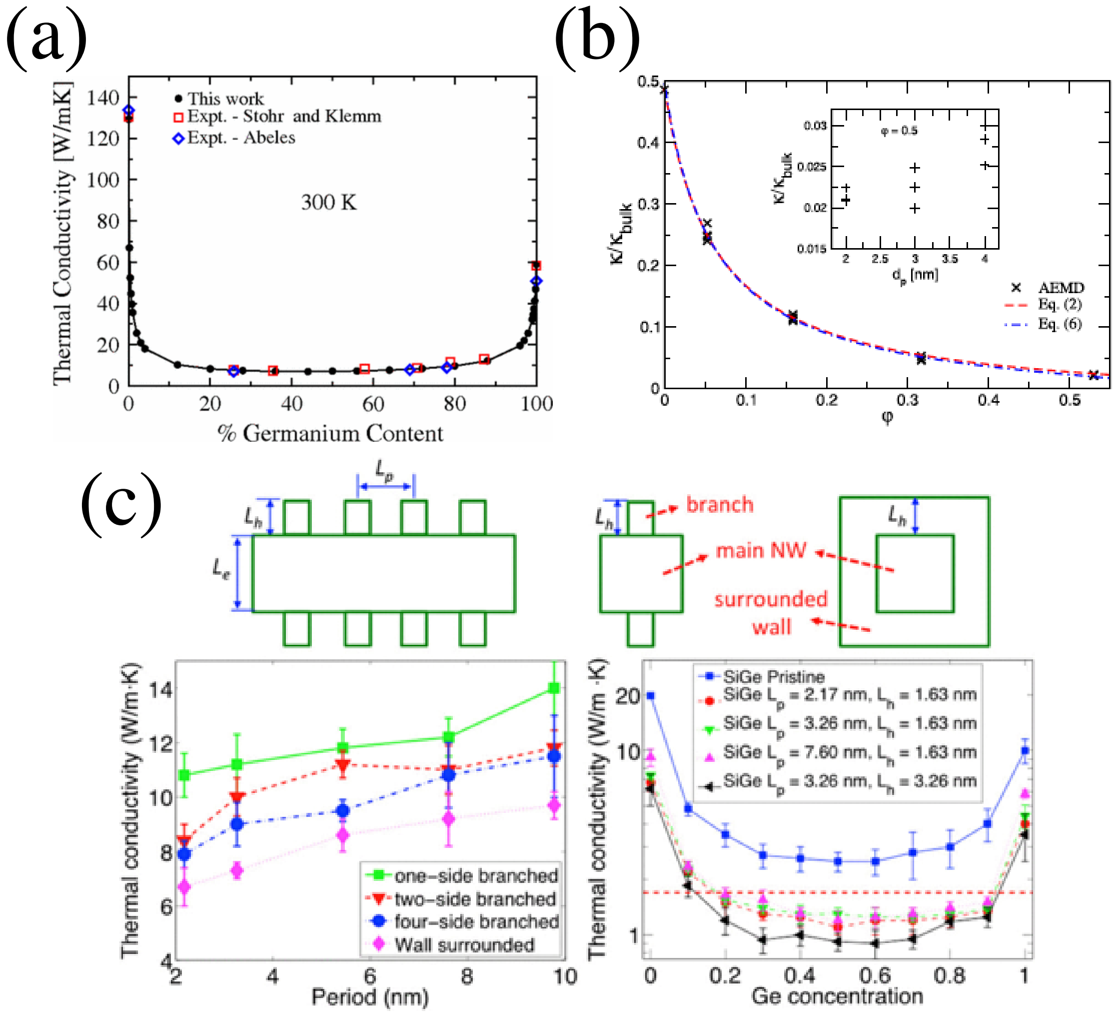

Figure 1 Dependence of the thermal conductivity with the concentration of defects in the case of (a) a SiGe alloy [41], (b) a porous Si nanowire [34], and a branched Si nanowires where resonant nanostructuring was combined with alloying [65]. Reprinted with permission from Ref. 41, copyright 2011 American Physical Society; Ref. 34, copyright 2016 American Institute of Physics; Ref. 65 copyright 2015 American Physical Society.

The effect of impurity scattering and boundary scattering have been successful combined in porous Si nanowires [34,40], in nanowires with a controlled level of isotopic composition [61,62,63] and in alloy nanowires [64]. For a combination of impurity scattering and phonon band structure engineering by means of superlattices (a topic discussed in the next section), on the other hand, one can see the work of Xiong et al. [65].

3. Superlattices

A superlattice (SL) is a lattice made by materials with different properties alternated in a periodic way and, consequently, separated by interfaces (*i.e.* boundaries). Therefore, SLs represent an ideal system for exploiting the different contributions to thermal transport at the nanoscale [66,67]. It is important to distinguish between two regimes depending on the phonon mean free path with respect to the superlattice period: phonon mean free path longer or shorter than the superlattice period.

If the phonon mean free path is smaller than the superlattice period, the thermal transport across the SL is defined by the series of the thermal resistance of the individual layers and the thermal resistance of the interfaces. Therefore, the total thermal resistance is given by the sum of the bulk and interfacial resistances of all the layers and the thermal conductivity decreases as the layers' thickness decreases [68,69,70,71].

If instead the phonon mean free path is longer than the superlattice period, both wave and particle-like phonon effects can be observed [72]. For this to happen it is necessary that the interfaces are as clean as possible[73], because it is in the ideal case of perfectly smooth interfaces that the phonons scatter specularly and can thus interfere coherently with their own reflections and modify their dispersion relations. These wave interference effects can lead to the formation of bandgaps and the modification of the density of states and group velocities [74,75,76,77,78,79]. The presence of a periodic superstructure can alter the lattice vibrational properties, *i.e.* the phonon spectra, of its constituent materials and can be designed in such a way that phonon transport in the SL structure is very different from that of their constituting materials [80,81].

Depending on the SL period with respect to the phonon mean free path, the wave-particle crossover is expected: for SL period smaller than the phonon mean free path wave theory applies and interference effects are expected to appear; for SL period larger than the phonon mean free path particle theory applies.

While single interfaces lead to diffusive phonon scattering, with consequent loss of phase information, a periodic repetition of interfaces can lead to constructive interference, with consequent coherent phonon transport [72]. Therefore, the interfaces of a SL can transmit phonons coherently.

Demonstrations of wave effects on macroscopic thermal transport quantities are less common than coherent wave transport of electrons and photons, despite their huge potential impact on thermal devices. Already at the end of the 70s, phonon transmission measurements through SLs at low temperatures unveiled phonon interference effects [80]. Later, inelastic light scattering experiments on SLs confirmed the formation of mini-bandgaps due to phonon interference in these structures [81,82]. Nevertheless, the coherence length of phonons probed with spectroscopic techniques is so short that the demonstration of wave interference effects in terms of macroscopic properties was rather elusive.

A linear dependence on the total SL length is the signature of coherent phonon transport. In the coherent regime, the phonon phase information is preserved at the interfaces of the SL, the superposition of the Bloch waves creates stop bands and effectively modifies the phononic band structure leading to a thermal conductivity that is linearly proportional to the total SL thickness. The experimental observation of coherent heat conduction has been reported only few years ago [83].

One of the important and long-standing predictions regarding thermal transport across superlattices is the existence of a minimum in the thermal conductivity as a function of the interface density, an indication of the crossover from particle-like to wave-like transport of phonons [75,84,85,86,87]. Measurements of the lattice thermal conductivity as a function of interface density in epitaxial oxide SLs provided the first unambiguous proof of wave-particle crossover, *i.e.* the transformation from the particle-like (incoherent) into wave-like (coherent) processes [88]. In the incoherent regime, each interface can be seen as a thermal resistance, the SL as a series of thermal resistances and, therefore, the lattice thermal conductivity decreases with interface density. In the coherent regime, the length of SL period is comparable to the coherence length of the phonons and the wave nature of phonons must be considered. Wave interference can lead to the formation of mini-bands. With increasing interface density, the number of mini-bands decreases, the spacing between them increases, and this leads to an increase in average phonon group velocity and thus in thermal conductivity. Experimentally, this wave-particle crossover (namely, coherent-incoherent crossover) turns into the existence of a minimum of the thermal conductivity as a function of interface density [88]. As an additional means for the engineering of thermal transport, the effect of boundary scattering can be further tuned with reducing the dimensionality of the structure by e.g. embedding the SL in nanowires [89,90,91,92] or embedding quantum dots in the SL layers [93,94].

Conclusions

We have discussed the importance of impurity and boundary scattering for the engineering of thermal transport for application ranging from thermal management to thermoelectric generation. As a particular type of boundary scattering, the importance of interface scattering is highlighted in the description of thermal transport in superlattices. Here, the transition from diffusive to coherent transport can be observed depending of the ration between phonon mean free path and superlattice period, due to phonon interference effects. By acting on these scattering mechanisms individually or by combining them suitable modifications of the phonon spectrum and of the phonon lifetimes can be achieved, thus enabling the design of materials with tailor made phononic properties.

Conflict of interest statement

Nothing declared.

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